Elementary processes and the mechanism of populating the working levels in a continuous-wave ion argon laser

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Electron excitation cross sections for the working levels of an ion argon laser are calculated in the Born-Coulomb approximation by taking into account autoionization of resonances. The radiative transition probabilities are determined concurrently. Semiempirical wave functions are employed in the calculations and deviation from LS coupling is taken into account. The population of the working levels in a CW laser is analyzed on the basis of these data. It is shown that the $4p\,^2D_{5/2}$ level is primarily populated as a result of the stepwise process $3p \to 3d\,^2F_{7/2} \to 4p\,^2D_{5/2}$, whereas the $4p\,^2p_{1/2,3/2}$ levels are mainly populated from the ground state of the ion. The results are compared with the experimental data.

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The main cause of population inversion in an argon ion laser (the short lifetimes of the lower working levels) is not subject to disagreement at the present time, but the same cannot be said concerning the mechanism whereby the working levels are populated. The main processes that are possible here in principle are well known (see, e.g., the reviews[1,2]), but a direct experimental determination of the contributions is as a rule a complicated experimental problem and in most cases has not yet been carried out. On the other hand, on the basis of indirect data and theoretical calculations, various authors arrive at different and still contradictory conclusions. The purpose of the present paper is to calculate, on the basis of the Born-Coulomb approximation, the probabilities of the elementary processes in an argon-discharge plasma and to analyze on the basis of these data the mechanism whereby the working levels of an argon ion laser are populated. We consider mainly transitions between doublet terms of the ion.

1. EXCITATION CROSS SECTIONS AND TRANSITION PROBABILITIES

In earlier studies[3,4] we calculated in the Born-Coulomb approximation the electron-excitation cross sections, averaged over the configurations, for a number of transitions in the argon ion, and used them for a preliminary analysis of the kinetics of the population of the configurations 4p and 4s. Although this approach, based on balance equations averaged over the configurations, was subsequently used in a number of works (see, e.g.,[5,6]), it is strictly speaking not fully valid, inasmuch as the rates of the radiative transitions in an argon laser greatly exceed the rates of population "mixing" within the configurations[7,8]. For a more accurate analysis, and in particular for a correct allowance for the influence of the metastable levels, it is necessary to know the probabilities of the transitions between individual levels of the fine structure. The corresponding calculations were made by us, as in the past, with semi-empirical wave functions on the basis of the Born-Coulomb approximation[9,10].

A. Coupling Scheme and Wave Functions of the Ion

In the calculation of the probabilities of the transitions between the fine structure, both radiative and via electron collisions, an important role is assumed by the correct choice of the coupling scheme. The coupling scheme for the argon ion was calculated many times[11-14] (the LS coupling is not suitable for the configurations of interest to us). We have used the data from Marantz's dissertation[11]. The spectrum and the structure of the terms of ArII have been well investigated[15,16].

A criterion (naturally, not an absolute one) of the quality of the wave functions can be the accuracy of the Einstein coefficients obtained with their aid. Table I lists the Einstein coefficients calculated in accordance with the coupling scheme of[11] for several lines of ArII in comparison with the published data, namely the results of calculations[11-14], experimental data[18-20], and data from the compilation review by Wiese et al.[17] It can be concluded from the table (and this agrees with the more detailed data[13,21]), that for the 4p–4s transitions the theoretical probabilities of the radiative transitions turn out to be quite close to the experimental ones. The small difference between the figures in the first and second columns of Table I is due to the difference between the values of the radial integrals, which were calculated in[11] with different radial wave functions. It should also be noted that calculations made by various authors[11-14] in the intermediate coupling scheme yield for the most part close results for the 4p–4s transitions (it is possible that the deviations in[12] are somewhat larger).

For transitions in which the levels of the configurations 3d and 4d take part, the situation is more complicated. Whereas for most transitions between the levels of the configurations 4p and 3d the theoretical values of the Einstein coefficients are in good agreement with the experimental data (this pertains primarily to intense lines), the difference can be quite appreciable for a number of transitions, especially for transitions with changes of the residue term (see[13]). In addition, the experimental lifetimes of certain levels of the configuration 3d[21] differ strongly (by 10 and more times) from the theoretical values[11]. Luyken[14] has proposed a possible explanation. According to[14], there is an appreciable configuration interaction in ArII, and the most strongly "mixed" configurations are $3s3p^6$, $3s^23p^4nd$ (n = 3, 4, . . .); on the other hand, for the configurations $3s^23p^44p$ and $3s^23p^44s$ the interaction is much less pronounced. The lifetimes obtained by Luyken for the levels of the 3d configuration are indeed in better agreement with the experimental data[21] than the data of Marantz[11]. In our present study we did not take into account the configuration interaction. The resultant error, generally speaking, is different for different levels. As will be shown later on, of greatest interest to us in configurations 3d and 4d are the levels $^2F_{7/2}$ and $^4F_{7/2}$. According to Table I, the probabilities of the transitions between these levels and the levels of the configuration 4p are calculated with an error of not more than 100%. Taking into account the error of the Born approximation itself for low electron energies (the "standard" error of the Born approximation is a factor on the order of two[10]) and the fact that for the terms of the configuration 4p the coupling scheme of[11] gives good results, it can be assumed that its use for the indicated two levels does not lead to a noticeable deterioration of the calculation accuracy.

B. Autoionization Resonances

In the calculations of electron excitation of ions, in addition to taking into account the distortion of the wave functions of the electron at large distances by the Coulomb field, it is necessary to take into consideration also the presence of autoionization states of an ion with multiplicity smaller by unity. In the case of ArII these are the autoionization states of the argon atom, and we are interested primarily in states produced upon simultaneous excitation of two electrons from the outer $3p^6$ shell. The autoionization states are arranged in series that are adjacent to the corresponding excited states of the ion, and can appear when the ion captures an electron that has a characteristic resonant energy for the given level (the width of the resonance is determined, as usual, by the lifetime of the corresponding level). Decay of such an autoionization state is possible either with emission of a photon (dielectron recombination) or in the manner of the Auger effect,[22] which corresponds to resonant elastic or inelastic scattering of the electron. In ions of low multiplicity, dielectron recombination proceeds predominantly from highly-excited autoionization states that occupy a very narrow energy band directly near the corresponding level of the ion. We shall not concern ourselves with this group of levels. The low-lying levels decay in the Auger manner, forming narrow resonances of the excitation cross sections.

The total contribution of the autoionization resonances to the excitation of the levels can be approximately taken into account within the framework of the Born-Coulomb approximation. The principles of the corresponding procedure were described by Beigman, Vainshtein, and Syunyaev[22]. They determined not the exact contour of the plot of the cross section against the electron energy, which has a complicated shape in the presence of resonances, but a smooth curve having the same area in other words, the cross section averaged over the individual resonances. Such a smooth curve can replace successfully the exact curve when the cross section is averaged over the velocity distribution of the electrons in the discharge. Moreover, a similar curve is obtained also when the excitation cross section is determined experimentally, if the scatter of the electron energies in the beam greatly exceeds the distances between the resonances.

In the calculations of the excitations cross sections of the levels of the $3p^44p$ configuration it was found essential to take into account the contribution of the autoionization states adjacent to the terms of the configuration $3p^44d$, and analogously in the case of $3p^44s$ and $3p^44p$. Figures 1, 2, and 3 show the cross sections for the excitation of the levels $4p^2D_{5/2}$, $4p^2P_{3/2}$, and $4p^2P_{1/2}$ from the ground state of ArII; the solid curves are the results of calculations, and the dashed curves are the experimental data of[23,24]. The experimental results given in[23,24] are not the cross section but the excitation functions of the spectral lines. To determine the cross sections it is necessary to know the branching factors of the corresponding ions, which were assumed to be 0.41, 0.77, and 0.78 respectively for the lines 4546, 4658, and 4880 Å (according to the data of Wiese et al.[17]). The steplike rises on the theoretical curves near the threshold constitute the smoothed-out (as indicated above) contributions of the autoionization resonances. The corresponding sections of the experimental curves are also smoothed out, since the electron-beam energy scatter was of the order of 2 eV; for the same reason, the experimental curves drop and go below the threshold. It can be concluded from Figs. 1–3 that the first maximum on the cross-section curves is due to the aforementioned autoionization resonances. Subsequently, the experimental and theoretical curves

TABLE I. Einstein coefficients for a number of transitions between the levels of the configurations 4s, 4d, 3d, and 4p (in units of 10^7 sec^{-1}).

Transition	Present paper	[11]	[12]	[13]	[14]* x	[14]* p	[17]	[18]	[19]	[20]
$4p\,^2D_{5/2}-4s\,^2P_{3/2}$	9.3	8.31	8.003	8.41	9.02	9.37	7.8	8.7	5.52	8.52
$4p\,^2D_{5/2}-4s\,^4P_{3/2}$	1.3	1.34	3.165	1.47	1.26	0.981	1.30	1.44	0.65	1.53
$4p\,^2P_{3/2}-4s\,^2P_{3/2}$	5.5	5.23	3.664	4.0	4.09	3.56	4.13	4.58	3.13	5.07
$4p\,^2P_{1/2}-4s\,^2P_{1/2}$	1.4	1.26		1.34	2.01	1.94	1.59	1.76	1.44	1.50
$4p\,^2P_{3/2}-4s\,^2P_{1/2}$	5.0	4.55	6.940	5.46	6.46	6.30	5.75	6.37	4.37	6.04
$4d\,^2F_{7/2}-4p\,^2D_{5/2}$	20						39			
$4d\,^4F_{7/2}-4p\,^4D_{5/2}$	19						27.7		21.8	
$4d\,^4D_{5/2}-4p\,^4P_{5/2}$	8.1						13.4		10.5	
$4p\,^2D'_{5/2}-3d\,^2F_{7/2}$	0.13			0.103	0.252	0.074			0.10	
$4p\,^4D_{5/2}-3d\,^4F_{7/2}$	1.2			1.06	1.406	1.08	1.13	1.25		

*In the left (x) and right (p) parts of this column we list the results of calculations with the matrix elements of the coordinate and momentum, respectively.

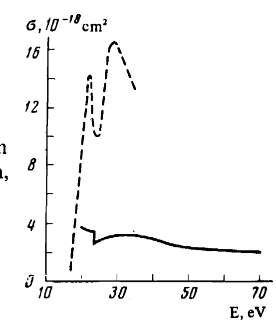

FIG. 1. Cross section for the excitation of the $4p^2D_{5/2}$ level: solid line—calculation, dashed—experiment [23,24].

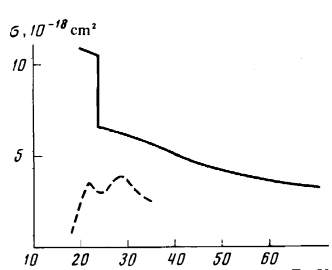

FIG. 2

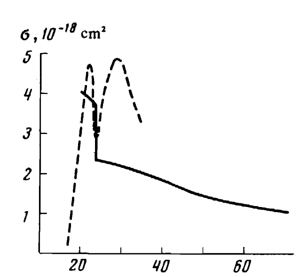

FIG. 3

FIG. 2. Excitation cross section of the level $4p^2P_{3/2}$: solid line—calculation, dashed—experiment [23,24].

FIG. 3. Excitation cross section of the $4p^2P_{1/2}$ level: solid line—calculation, dashed—experiment [23,24].

diverge, and whereas for the $4p\,^2P_{1/2}$ and $4p\,^2P_{3/2}$ levels the deviation from the experimental data does not exceed on the whole the aforementioned "standard" error, the discrepancy is fivefold for the $4p\,^2D_{5/2}$ level.

It is difficult to judge at present what improvements are primarily needed in the calculation procedure for the considered transitions. On the other hand, account must also be taken of the possible errors in the experiment, which technically is a rather complicated task and so far, to our knowledge, has never been repeated. It is also important to note that at the present time there are experimental data for the excitation functions of a number of levels of the ion only from the ground state. These data are insufficient for a complete analysis of the mechanism that produces inversion in the laser.

Calculations of the cross section for electron-impact excitation of the levels of the configuration 4p of ArII from the ground state of the ion were carried out also by Brandy and Koster[25,26] by the distorted-wave method with allowance for exchange. The contributions of the autoionization resonances were not taken into account in these papers. In[26] are given the cross sections for the excitation from the ground state $3p^5\,^2P_{3/2}$ of ArII. For the $^2D_{5/2}$ and $^2P_{1/2}$ levels they do not differ greatly from our data near the threshold[1)], but reveal a characteristic dependence on the electron energy: the cross sections approximately double when the energy changes from 2 to 5 Ry. For the $^2P_{3/2}$ level, the cross section near the threshold, according to[26], is approximately one-sixth of our cross section[1)] (in this case the experimental data[23,24] are intermediate in value). With increasing energy, this cross section, according to[26], also increases. We present for comparison the relative values of the excitation rate constants for electron temperatures typical of lasers. If we take as unity the corresponding value for the $^2P_{1/2}$ level, then we obtain for the $^2P_{3/2}$ level 0.31, 0.77, and 2.8 if we use respectively the data of[24,25] and our data[1)]. For the $^2D_{5/2}$ the corresponding figures are 1.2, 3.2, and 1.0 (for the same sources). These data show once more that from the first Born approximation and its modifications one cannot expect the higher accuracy than within the limits of the already mentioned "standard error." Vladimirova et al.[27], on the basis of measurements of the radiation intensity of the discharge in the presence and in the absence of generation, estimated the de-activation rate constants of the upper laser levels in collisions with electrons under discharge conditions typical of lasers. For the doublet terms of the 4p configuration they obtained a value 6×10^{-7} cm^3 sec^{-1}. Our data give 9.3×10^{-7} cm^3 sec^{-1} (averaged over three levels: $4p\,^2P_{1/2}$, $4p\,^2P_{3/2}$, and $4p\,^2D_{5/2}$). A similar summary of the rate constants of the elementary processes is presented in Tables II and III below.

2. KINETICS OF LEVEL POPULATION

A. Level Scheme and Transition Probabilities

To calculate the laser level populations in the absence of generation, we used a system of balance equations that included electronic impacts of the first and second kind and radiative transitions. The two-electron transitions

$$\mathrm{Ar}+e\to(\mathrm{Ar}^+)^*+2e,$$

in which the atom is simultaneously ionized and excited from the ground state[28] do not play an essential role in a cw laser, as was demonstrated by direct measurements of Bennett and co-workers[29]. The same conclusion follows from a comparison of the drift velocities of the atoms and ions in a discharge with the Doppler shift of the generation lines in an argon ion ring laser[30].

An analysis of the cross sections and the radiative-transition probabilities has made it possible to select processes that must be taken into account primarily in the analysis of the kinetics of the population of the laser levels. We note immediately that the level $4p\,^2D_{5/2}$ is coupled by intense transitions to the $^2F_{7/2}$ terms of the configurations 3d and 4d, whereas the influence of the metastable and slowly-decaying levels of the same configurations to the levels $4p\,^2P_{1/2}$ and $4p\,^2P_{3/2}$ are much smaller. Figure 4 shows the level scheme used to calculate the populations of the terms $4p\,^2P_{1/2}$, $4p\,^2P_{3/2}$, and $4p\,^2D_{5/2}$. Certain groups of levels and even configurations could be combined in this case into the blocks represented by the shaded rectangles. In order not to clutter up the figure, the arrows indicate the principal transitions that are taken into account in the determination of the population of the $4p\,^2D_{5/2}$ level. Wavy lines denote radiative transitions. Impacts of the second kind were taken into account automatically. The block 5g was included in the scheme as an effective sink for the levels of the configuration 4f. Since this block is coupled with the working laser levels of interest to us by a long chain of transitions, an exact description of the de-activation processes is immaterial, and the latter were artificially replaced by an intensive radiative transition to the ground state (designated by a dashed line in the figure); the probability of this transition, of course, is in no way connected with the real transition probability.

The probabilities of the elementary processes used in the calculation of the populations in accordance with the scheme of Fig. 4 are listed in Table II. For the excitation rate constants we used a two-parameter analytic approximation[9]

$$\langle v\sigma_{01}\rangle=10^{-8}\left(\frac{\mathrm{Ry}}{\Delta E}\frac{E_1}{E_0}\right)^{1/2}A\frac{F(\beta)}{\beta+\chi}e^{-\beta}. \tag{1}$$

Here E_0 and E_1 are the level energies reckoned from the ionization boundary; $\Delta E = E_0 - E_1$; Ry is the Rydberg energy unit; $\beta = \Delta E/kT_e$ (k is the Boltzmann constant); A (which must not be confused with the Einstein coefficient) and χ are approximation parameters, with A so normalized that $\langle v\sigma\rangle$ has the dimension cm^{3}sec^{-1}; for most transitions in the table we have $F(\beta) = (\beta + 1)\sqrt{\beta}$, and for the remaining ones (the corresponding numbers are underscored) we have $F(\beta) = \beta^{3/2}$. The accuracy of

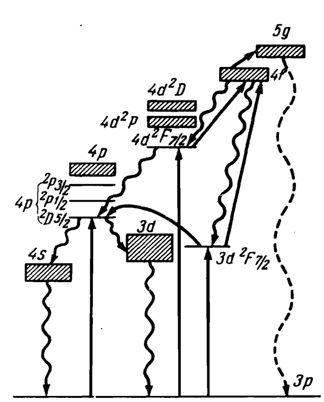

FIG. 4. ArII term scheme.

the approximation in the temperature range from 2 to 9 eV lies as a rule in a range of 10%. The table lists the values of g_0A, where g_0 is the statistical weight of the lower level (or of the group of levels). The rate constant of the inverse transition $1 \rightarrow 0$ differs in this case from (1) only in the absence of the factor $e^{-\beta}$. In the column headed gf are listed the oscillator strengths used in the calculations (again, more accurately) quantities multiplied by the statistical weights of the levels). In the calculation of the populations of the 4s terms we took into account a smaller number of configurations. The corresponding transition probabilities are gathered in Table III.

B. Calculation Results. Comparison with Experiment

The system of balance equations was solved numerically with the aid of the special program developed by L. A. Vaĭnshteĭn. Figure 5 shows the specific populations of the laser levels (i.e., the populations divided by the statistical weight of the level) as functions of the electron temperature at a constant electron concentration[2)] 4×10^{13} cm^{-3}.

TABLE II. Probabilities of the elementary processes for the calculation of the 4p configuration level populations

Transition	gf	g_0A	χ	Transition	gf	g_0A	χ
$3p-4s$	1.24			$3d-4p\,{}^2P_{3/2}$	0.489	6.82	0.412
$3p-3d\,{}^2F_{7/2}$		62.3	0.001	$3d-4p$	0.477	3.58	0.412
$3p-3d$	12.8			$3d-4d\,{}^2P$		13.2	0.579
$3p-4p\,{}^2D_{5/2}$		6.65	0.015	$3d-4d\,{}^2D$		22	0.579
$3p-4p\,{}^2P_{1/2}$		7.66	0.015	$3d-4f$	4.39	68.8	0.171
$3p-4p\,{}^2P_{3/2}$		21.2	0.01	$3d\,{}^2F_{7/2}-4f$	1.6	25	0.171
$3p-4d\,{}^2F_{7/2}$		57.5	0.15	$4p\,{}^2D_{5/2}-4d\,{}^2F_{7/2}$	3.04	25	0.16
$3p-4d\,{}^2P$	0.59	76.8	0.285	$4p\,{}^2D_{5/2}-4d\,{}^2D$	0.582	4.8	0.16
$3p-4d\,{}^2D$	1.77	230	0.285	$4p\,{}^2P_{1/2}-4d\,{}^2P$	0.323	2.67	0.16
$4s-4p\,{}^2D_{5/2}$	2.29	24.5	0.303	$4p\,{}^2P_{1/2}-4d\,{}^2D$	0.97	8.0	0.16
$4s-4p\,{}^2P_{1/2}$	0.763	8.17	0.303	$4p\,{}^2P_{3/2}-4d\,{}^2P$	0.647	5.33	0.16
$4s-4p\,{}^2P_{3/2}$	1.53	16.3	0.303	$4p\,{}^2P_{3/2}-4d\,{}^2D$	1.94	16	0.16
$4s-4p$	2.29	24.5	0.303	$4p-4d\,{}^2P$	1.32	11.2	0.16
$3d\,{}^2F_{7/2}-4p\,{}^2D_{5/2}$	0.477	6.65	0.412	$4p-4d\,{}^2D$	0.388	3.2	0.16
$3d\,{}^2F_{7/2}-4p$	0.11	1.53	0.412	$4d\,{}^2F_{7/2}-4f$	5.72	11.1	0.001
$3d\,{}^2F_{7/2}-4d\,{}^2F_{7/2}$		15.0	0.579	$4d\,{}^2P-4f$	4.29	8.34	0.001
$3d-4p\,{}^2D_{5/2}$	0.257	10.2	0.412	$4d\,{}^2D-4f$	7.15	13.9	0.001
$3d-4p\,{}^2P_{1/2}$	0.244	3.41	0.412	$4f-5g$		144	0.038

TABLE III. Probabilities of the elementary processes for the calculation of the 4s configuration level populations*

Transition	gf	g_0A	χ	Transition	gf	g_0A	χ
$3p-4s\,{}^2P_{3/2}$	0.81	9.78	0.021	$4s\,{}^2P_{3/2}-4p'\,{}^2P_{3/2}$	0.115	1.23	0.303
$3p-4s\,{}^2P_{1/2}$	0.41	5.25	0.034	$4s\,{}^2P_{3/2}-4p'\,{}^2P_{1/2}$	0.0665	0.71	0.303
$3p-3d$	12.8			$4s\,{}^2P_{1/2}-4p\,{}^2D_{3/2}$	0.77	8.23	0.303
$3p-4p\,{}^2D_{5/2}$	0.00465	6.65	0.015	$4s\,{}^2P_{1/2}-4p\,{}^2P_{1/2}$	0.102	1.09	0.303
$3p-4p\,{}^2D_{3/2}$		3.18	0.034	$4s\,{}^2P_{1/2}-4p\,{}^2P_{3/2}$	0.678	7.25	0.303
$3p-4p\,{}^2P_{1/2}$		7.66	0.015	$4s\,{}^2P_{1/2}-4p\,{}^2S_{1/2}$	0.596	6.37	0.303
$3p-4p\,{}^2P_{3/2}$		21.2	0.011	$4s\,{}^2P_{1/2}-4p'\,{}^2P_{3/2}$	0.0332	0.355	0.303
$3p-4p\,{}^2S_{1/2}$		4.37	0.062	$4s\,{}^2P_{1/2}-4p'\,{}^2P_{1/2}$	0.063	0.674	0.303
$3p-4p'\,{}^2P_{3/2}$	0.0304	39.7	0.556	$3d-4p\,{}^2D_{5/2}$	0.733	10.2	0.412
$3p-4p'\,{}^2P_{1/2}$	0.0152	21.1	0.556	$3d-4p\,{}^2D_{3/2}$	0.489	6.82	0.412
$4s\,{}^2P_{3/2}-4p\,{}^2D_{5/2}$	2.0	21.4	0.303	$3d-4p\,{}^2P_{1/2}$	0.244	3.41	0.412
$4s\,{}^2P_{3/2}-4p\,{}^2D_{3/2}$	0.676	7.23	0.303	$3d-4p\,{}^2P_{3/2}$	0.489	6.82	0.412
$4s\,{}^2P_{3/2}-4p\,{}^2P_{1/2}$	0.543	5.81	0.303	$3d-4p\,{}^2S_{1/2}$	0.244	3.41	0.412
$4s\,{}^2P_{3/2}-4p\,{}^2P_{3/2}$	0.677	7.24	0.303	$3d-4p'\,{}^2P_{3/2}$	0.489	6.82	0.412
$4s\,{}^2P_{3/2}-4p\,{}^2S_{1/2}$	0.151	1.62	0.303	$3d-4p'\,{}^2P_{1/2}$	0.244	3.41	0.412

*For certain 3p–4p transitions the table indicates the oscillator strengths. These are fictitious quantities, with values chosen such as to take correct account of the decays from the corresponding 4p states to those 4s configuration levels not included in the calculation scheme.

Figure 6 gives the dependence of the level populations on the electron concentration n_e. It shows the ratios of the level populations to the square of the electron concentration at constant temperatures. The figure shows relative units, unity for each level being the corresponding value at $n_e = 4 \times 10^{13}$ cm^{-3}. The curves for the levels $4p^2P_{1/2}$ and $4p^2P_{3/2}$ differed by not more than 10–15%, so that one common curve is drawn for them. With the same accuracy, the curves of Fig. 6 are suitable for the entire considered temperature range.

It follows from Fig. 6 that the dependence of the level populations of the 4s concentration on the electron concentration is nearly quadratic, thus indicating that their decay is predominantly radiative. On the other hand, for the 4p levels at electron concentrations on the order of several times 10^{13} cm^{-3}, impact deactivation already comes into play. It must be borne in mind here, however, that in the calculations we did not take into account the dragging of the resonant radiation of ArII. Since the decay of the doublet 4s levels proceeds mainly on account of resonant radiative transitions, the not too strong dragging of the radiation can be taken into account by simply increasing the calculated values of the populations by as many times as the effective lifetime of the levels is increased. Under typical discharge conditions in a tube of 2 mm diameter, the quantity k_0R (k_0 is the absorption coefficient at the center of the resonance line and R is the radius of the tube) turns out to be of the order of unity. It must be recognized, however, that calculation of the effective lifetime by the Holstein-Biberman theory[32] will be incorrect and can be used only as an estimate (in fact, an overestimate), since both the ion concentration and the ion velocity distribution function are not constant over the tube cross section (see[33]).

To estimate the role of different channels in the population of the upper laser levels, we determined

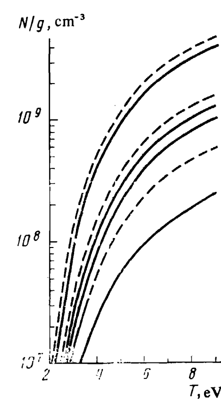

FIG. 5

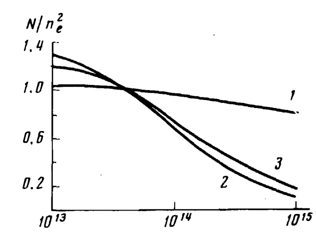

FIG. 6

FIG. 5. Dependence of the ArII level populations on the electron temperature at $N_e = 4 \times 10^{13}$ cm^{-3}: curve 1 for $4p^2D_{5/2}$; curve 2 for $4p^2P_{1/2}$; curve 3 for $4p^2P_{3/2}$; curve 4 for $4s^2P_{1/2,3/2}$. Solid curves—results of calculations using only the theoretical cross section; dashed—obtained by replacing the excitation cross sections of the 4p levels from the ground state of ARII by the experimental values [23,24] (the cascades from the 4d levels are not taken into account, since they are already included in the experimental excitation functions). The populations of the levels $4s^2P_{1/2}$ and $4s^2P_{3/2}$ practically coincide (a difference of several percent) and are therefore represented by a single common curve.

FIG. 6. Dependence of the level populations on the electron concentration. Details in the text. Curves 1, 2, and 3 are for 4s, $4p^2D_{5/2}$, and $4p^2P$.

their relative contributions to the total pumping of the corresponding levels. These estimates were carried out at $n_e = 4 \times 10^{13}$ cm^{-3} and $T_e = 4$ eV. It turned out that the $4p^2D_{5/2}$ level is populated mainly via the stepwise process $3p \to 3d^2F_{7/2} \to 4p^2D_{5/2}$ (approximately 80% of the total pumping). This is precisely why the dashed and solid curves for this level in Fig. 5 differ so little, in spite of the considerable difference between the measured and calculated cross section for the excitation of the $4p^2D_{5/2}$ level from the ground state of the ion (Fig. 1). In view of a number of technical difficulties, no complete calculations were carried out for the $4p^4D_{5/2}$ level. Only estimates were made, according to which the population of this level also has a predominantly stepwise character, but this time as a result of the process $3p \to 3d^4F_{7/2} \to 4p^4D_{5/2}$. The levels $4p^2P_{1/2}$ and $4p^2P_{3/2}$ are populated predominantly from the ground state. Cascade transitions from higher levels of the 4d configuration make contributions 14, 5, and 6% to the population of the levels $4p^2D_{5/2}$, $4p^2P_{3/2}$, and $4p^2P_{1/2}$, respectively (according to measurements by Rudko and Tang[34], these values are 27.4%, 14.5%, and 3.7%). The deduced stepwise character of the excitation of the levels $^2D_{5/2}$ and $^4D_{5/2}$ in an argon ion laser agrees with the conclusion of Gur'ev et al.[35] This hypothesis was first advanced at the Bell Telephone Laboratories.[3)] A similar point of view is advanced also in[36,37]; there, however, the role of the principal intermediate link in the stepwise process is assigned to quartet terms of the 4s configuration (without sufficient justification, in our opinion). Furthermore, in the latter papers no difference is made between the mechanisms for the population of the different levels of the 4p configuration. On the other hand, a number of workers (including Brandy[25]) do not note the role of the stepwise processes, and attribute the pumping of the upper laser levels mainly to excitation from the ground state of the ion.

For a comparison of the calculated and measured values of the level populations it is necessary to know the concentration and temperature of the electrons in the discharge. The diagnostics of the plasma of an argon laser is a complicated experimental task, and direct measurement methods were developed for it only recently[38,39]. It has turned out here that the experimental data are in general satisfactorily described by the Tonks-Langmuir-Klarfeld theory[40]. To determine the temperature and concentration of the electrons we have therefore used mainly this theory[1,41]. The gas concentration was determined with allowance for its being forced out from the discharge tube by heating[42]. The results of the calculations are compared with the experimental data in Table IV. It is seen from the table that for the population of the $4p^2D_{5/2}$ level the agreement with the experimental data is quite satisfactory. An appreciable discrepancy was obtained only with the results of Rudko and Tang[34]. It is possible that calculation by the procedure of[41,1] underestimates in this case the electron concentration.

For the levels $4p^2P_{3/2}$ and $4p^2P_{1/2}$, the deviation from the experimental values is larger than for $4p^2D_{5/2}$, and an even greater difference appears for the levels of the 4s configuration (up to 10 times). The discrepancy between the experiment and the results of the calculations, besides the possible measurement errors, is due also to inaccuracies in the calculations of the cross sections and to errors in the diagnostics of the discharge plasma. The influence of the latter is eliminated to a considerable degree by changing over to relative quantities. It turns out here, according to the data of various authors, that the specific population of the $4p^2D_{5/2}$ level is larger than those of the levels $4p^2P_{1/2}$ and $4p^2P_{3/2}$ by a factor 1.1–2.6. According to the calculation, this value is close to 4. With respect to the 4s levels, it must be borne in mind that no account of the dragging of the resonant radiation was taken in the calculation (see above). In addition, the populations of these levels were determined not by radiation directly (for which purpose it would be necessary to carry out measurements in the vacuum ultraviolet), but by using the gain in the laser transitions. This could lead to additional errors, inasmuch as no account was taken in[45] and[43] of the transverse and longitudinal (cf.[46]) inhomogeneities of the discharge:

CONCLUSION

Calculations of the probabilities of the elementary processes in ArII are complicated by the noticeable deviation from the LS coupling and by the presence of a configuration interaction in the ion. Since these phenomena are more strongly pronounced for heavier inert gases, the analogous calculations for them are even more complicated and less reliable. In view of the known shortcomings of the theory, it would be quite desirable to obtain additional more detailed experimental data on the cross sections.

TABLE IV. Populations of laser levels of argon ion. Comparison with experiment.

R, mm	P, Torr	j, A/cm^2	T_e, eV	n_e, 10^{13} cm^{-3}	N/g, 10^{3} cm^{-3}													Reference
					$4p\,^2D_{5/2}$			$4p\,^2P_{3/2}$			$4p\,^2P_{1/2}$			$4s\,^2P_{3/2}$		$4s\,^2P_{1/2}$		
					I	II	III	I	II	III	I	II	III	I	II	I	II	
1.25	0.45	200	3.8	4.5	8.9	4.1	5.1	3.4	1.2	0.59	5.9	0.89	1.5	2.4	0.24	1.0	0.24	[43]
1.0	0.3	159	4.3	2.1	8.0	1.8	2.3	7.0	0.51	0.24	7.0	0.39	0.65					[34]
1.0	0.6	159	3.7	3.4	3.1	2.2	2.7	2.3	0.73	0.34	2.3	0.55	0.91					[44]
1.0	0.2	223	4.8	2.9	2.7	4.8	6.0	2.5	1.4	0.67								[28]
1.2	0.5	265	3.9	12 *	13.7	23	28											[35]
1.4	0.3	160	{ 3.4 *	4.3 *	4.1	2.2	2.7							1.6	0.13			[45]
			4.1	3.6	4.1	3.8	4.8							1.6	0.21			
2.4	0.3	200	4.0	5.8	9.0	7.8	9.9							3.8	0.49			[45]

Note. Roman numerals denote: I—experimental data, II—results of calculations, and III—results of calculations using the cross sections measured by Zapesochnyĭ et al. [23,24] The asterisks mark the experimental values obtained simultaneously with the level populations.

Inasmuch as the curves of Fig. 6 depend very little on the electron temperature (at T_e = 2–9 eV), the plots of Figs. 5 and 6 can be used to determine the populations of the levels represented on them in the entire considered range of electron temperatures and concentrations. It turns out here that, according to calculation, the population inversion in the laser transitions is preserved with large margin in a wide range of n_e. However, at large electron concentrations it is necessary to introduce in the calculations, as already mentioned, a correction for the dragging of the resonant radiation. This question was discussed many times[1,46,47], but has not yet been satisfactorily answered. This in turn hinders the explanation of the reason for the nonmonotonic variation of the generation power with increasing discharge current. Among the various causes, besides dragging of the resonant radiation and impacts of the second kind, we have indicated here the possibility of nonmonotonic variation of the concentration of the electrons with increasing current[48]. Nonetheless, this question has not yet been finally answered.

In spite of the shortcomings of the method used to calculate the electron-excitation cross sections, the results, together with the aggregate of the experimental data, give grounds for assuming that the pumping mechanism of laser levels in a cw argon ion laser has been in the main explained, although many questions (including the cause for the breakdown of the lasing) still call for refinement.

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1)It should be borne in mind that our data, in contrast to [26], pertain to excitation from both fine structure levels of the ArII ground term $3p^5\ ^2p$.

2)Calculation errors have crept into the previously published data [31].

3)R. Miller, oral communication.

4)Formula (7) of [49] and the notation in it contain misprints. These have been corrected in the present text (formula (1) and beyond).

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